PROCESS FOR PRODUCING CARBONACEAOUS SOLID MATERIALS AND HYDROGEN-RICH GASES

The present invention relates to methods for producing solid carbonaceous materials and hydrogen-rich gases, methods of combustion, reactors for the same and use of such reactors.

Natural gas and several of its derivatives are most commonly used for energy purposes, by conversion to heat and electricity. With few exceptions, this conversion is started with a combustion process, where the so-called heating value of the fuel gas is released by converting the hydrogen and the carbon in the fuel gas into water (vapour) and CO_2. The oxygen necessary to perform this reaction is derived from the atmosphere which consists of 21% oxygen and 79% nitrogen.

Natural gas normally contains more than 80% methane and thus combustion may be represented by the overall reaction equation:

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$$
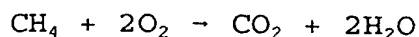
This combustion provides a heating value of 50000kJ/kg of methane. The combustion of for example CH_4 includes as many as 400-500 elementary reactions. However, only a few of these lead to a significant energy release in terms of heat.

Viewed in a simplistic way, the carbon is converted during combustion as follows:

$$C \rightarrow CO \rightarrow CO_2$$

If oxidation is divided into two steps as indicated above, the heat related to the formation of CO is only moderate causing only a small temperature increase. However, a large temperature increase is associated with the formation of CO_2. Thus the main heat release in hydrocarbon flames is associated with the formation of CO_2

and H_2O.

When combustion of hydrocarbons take place a lot of intermediate species are formed, most of which are combusted before the products leave the flame so that in general the final products which result are in the form of CO_2 and H_2O.

In flames, soot may also be formed. The formation of soot takes place on the fuel rich side of the flame and results as a consequence of heating the fuel. Soot is a hydrocarbon with 1-3% by weight of hydrogen and is a polybenzenoid molecule. It has been suggested that it may have a structure similar to $C_{40}H_{16}$.

Soot formation from the gas phase includes a dehydrogenation process, similar to cracking and/or pyrolytic decomposition, which probably produces the product C_2H_2 before being built up into a larger molecule by virtue of several thousand steps. However, in macroscopic terms, the soot formation process may be viewed as a two step process, namely the formation of a nucleus and then particle formation. The soot particles look like small spheres that may be agglomerated into clusters like grapes. The individual spheres have a diameter in the order of 10-100 nanometres.

In normal combustion of a hydrocarbon fuel, the fuel is introduced into the combustion chambers or burner systems in the form of jets or fuel sprays, or in other cases as a mixture between this fuel and the oxidizer, which is usually oxygen from the air. Usually two types of flames may be formed depending on the types of burner systems which are employed.

Diffusion flames occur when the fuel and oxygen are mixed on both the macro and micro, but not molecular scale through diffusive processes. Since combustion only occurs when reactants are mixed on a molecular scale, this gives rise to laminar or turbulent flames. Thus, the so-called thermal reactions resulting from the high temperatures of the laminar and/or turbulent flames take

place only in localized regions such as thin sheets, vortex structures or similar which lie at the interface between the fuel and the oxygen. Generally the so-called thermal reactions at these sites are very fast and the temperature in the reaction zone is very high.

Pre-mixed flames occur when the fuel and oxygen are pre-mixed on a molecular scale. Generally a propagating flame is formed which has associated with it a flame velocity which refers to the propagating velocity of the flame. Such flames may be fuel-rich or fuel-lean. Many combustion devices utilize lean pre-mixed combustion as the reaction temperature is low thus leading to a reduced formation of NO_x (which is formed from N_2 in air) since reactions leading to NO_x formation are generally slow and strongly temperature dependent. In both of the above types of flames the same chemical reactions occur, albeit at different sites, but the influence on the products of the different reactions may vary.

In an ordinary combustor or burner, only a small fraction of the carbon is converted into soot and the soot burns away before the product leaves the flame. In some cases such as in lean premixed flames no soot is formed. In fires with big diffusion flames, where very fuel-rich structures are heated, large amounts of soot are produced. Sometimes 10-20% of the carbon in the fuel forms as soot which is not burnt off.

However, a major disadvantage with this combustion is the production of CO_2 which is detrimental to the environment. Furthermore, the soot-like material which forms, which has potential uses in a number of industrial applications is lost.

US-5527518 (Kvaerner Engineering AS) describes a method of pyrolytic decomposition of natural gas (or methane) which results in the production of a hydrogen-rich gas by the use of a plasma torch which heats the hydrocarbon fuel to about 1600°C. Instead of the production of CO_2, carbon black, which may contain some

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hydrogen, is produced. This reaction is performed in the
absence of oxygen except in reactions to modify the
properties of the carbon black which has already been
produced.

It has now surprisingly been found however, that at
moderate detriment to the heating value per kg of fuel
gas, harmful emissions of CO_2 may be reduced by conversion
of the carbon content of the fuel into a solid form of
carbon which has useful industrial applications.

This is achieved by manipulation of the above
mentioned reaction scheme by using a controlled amount of
oxygen in the system and by using a combustion process,
which may be separated into 2 separate stages, coupled
with pyrolytic decomposition.

By controlling the amount of oxygen in the system,
the equation may be rewritten as:

$$CH_4 + O_2 \rightarrow C + 2H_2O$$
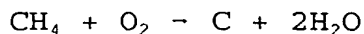
In the above reaction, while hydrogen is converted
to water, carbon is decomposed from the fuel and forms as
a solid material similar to soot. As a consequence, very
little or no CO_2 forms. Such processes are achievable and
still retain more than 50% of the original heating value.
However in most cases it is realistic to expect that some
carbon will react to form CO_2 and some will produce solid
carbon, depending on the operating conditions and the
reactor vessel design. Thus for example, a practical,
readily or easily achievable reaction may be written as:

$$CH_4 + 1.5O_2 \rightarrow 0.5C + 0.5CO_2 + 2H_2O$$
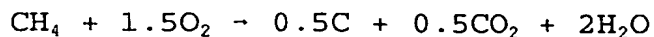
This reaction would provide a heating value of
40000kJ/kg fuel gas, produce 50% less CO_2 and produce
solid carbon at an amount of 0.375kg/kg methane. Thus a
significant reduction in CO_2 emissions may be achieved
with only a 20% reduction in heating value.

In the above equations sufficient oxygen is used to allow all the hydrogen in the reaction to be converted to H_2O. If however the levels of oxidizer are even further limited, some of the hydrogen in the reaction will be converted to H_2. Thus for example using methane as the hydrocarbon fuel (comprising both the feedstock and flame fuel) and limiting the amount of oxidizer, the equation may for example be rewritten as:

$$CH_4 + 0.1\ O_2 \rightarrow C + 1.8\ H_2 + 0.2\ H_2O$$
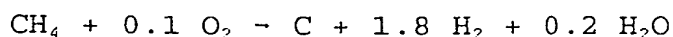
Alternatively if hydrogen is used as the flame fuel the above equation may be rewritten as:

$$CH_4 + 0.2\ H_2 + 0.1\ O_2 \rightarrow C + 2H_2 + 0.2\ H_2O$$
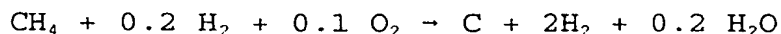
In both of the above equations the reactions produce enough heat to facilitate heating of the feedstock and consequent decomposition. Thus an oxidizer, such as oxygen, in stochiometric amounts such as 0.2 or less relative to the amount of carbon in the system may be employed. This is equivalent to 5% or less of the total amount of oxidizer required for complete combustion.

During the course of the above described reaction, unlike known reactions, significant amounts of solid carbon and hydrogen-rich gas are produced. This is achieved through incomplete combustion of the feedstock by control of the amount of oxidizer, e.g. pure oxygen, which is introduced into the system thus preventing continued combustion such that these products may be collected. The hydrogen-rich gas may then be combusted immediately or after collection to provide an energy source.

Thus, viewed from one aspect, the present invention provides a method of producing solid carbonaceous C_xH_y, wherein x and y are the average number of carbon and hydrogen atoms respectively and the ratio of x:y is

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greater than 2.5:1, wherein a hydrocarbon fuel
(comprising bulk feedstock fuel and flame fuel) is heated
with oxygen at a C:O ratio greater than 1:3, preferably
1:2 (e.g. greater than or equal to 1:1, e.g. 1:0.4,
particularly preferably 1:0.2) to a temperature of at
least about 1000°C whereby to effect incomplete
combustion and pyrolytic decomposition of said
hydrocarbon fuel. In this system, the flame fuel is
combusted and the combustion products which are produced
are used to heat the bulk feedstock fuel to a temperature
of at least about 1000°C to achieve pyrolytic
decomposition.

As referred to herein, the C:O ratio refers to the
stoichiometric ratio of carbon and oxygen atoms and a
greater ratio refers to a ratio containing relatively
more C.

Preferably the value of x in the x:y ratio exceeds
40, leading to H_y of less than 1-3% by mass in C_xH_y.

As referred to herein, "hydrocarbon fuel" also
referred to as "fuel source" or "fuel gas", refers to any
commercially or readily available hydrocarbons which are
gaseous, liquid or solid (such as natural coal) at a
temperature of 20°C and a pressure of 1 bar, preferably
one or more gaseous hydrocarbons, and which are
susceptible to pyrolytic decomposition and/or combustion.
This includes the fuel (preferably a gas) used to create
the flame (referred to herein as the "flame fuel" or
"flame gas") which may be the same or different to the
bulk feedstock fuel. Particularly preferred is natural
gas which is composed mainly of methane. Other
appropriate hydrocarbons, particularly for use as flame
fuels, include hydrogen (H_2), acetylene (C_2H_2) and propane
(C_3H_8).

If solid fuels are to be used, it will be
appreciated that appropriate particulation to a powder
form will be necessary before introduction into the
reactor. Appropriate techniques for preparing such
powders and for introduction into reactors are well known

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in coal driven power plants in which coal powder is blown into the reactor. In the present invention the gas carrier of such powders may conveniently be the exhaust gases produced by the method of the invention.

Conveniently, flame fuels are the preferred fuels described above or any other combustible fuels that can create high temperatures when combusted with air or oxygen. "Flame fuel", "hydrocarbon fuel" and "bulk feedstock fuel" as used herein refers to the hydrocarbon component in additional to any associated trace carrier gas, e.g. air, oxygen, nitrogen, CO_2 etc., where present. Trace amounts of oxygen which may be present with the hydrocarbon fuel, such as in air do not constitute the oxidizer required for performance of the invention. This oxidizer is supplied in a controlled and precise manner.

Thus, the present method allows the production of a solid carbonaceous product containing 20% or more, preferably 50% or more, for example at least 80% of the carbon introduced into the system, by subjecting the hydrocarbon fuel to thermal reactions with limited combustion achieved by the use of restricted amounts of an oxidizer. Combustion of the hydrocarbons of a portion of the hydrocarbon fuel (ie. that portion which is the flame gas, which may be the same as or different, and may be mixed with or separate to the remaining feedstock) produces hot combustion products which then serve to heat the remaining feedstock (to temperatures of at least 1000°C) producing the reactions required for solid carbon/hydrogen-rich gas production.

Preferably the method is performed by pre-heating the feedstock fuel (fuel source) to between 600 and 800°C, for example approximately 600°C and passing said fuel source into a reactor. By raising the temperature of the fuel source, the hydrocarbon reaches a temperature close to the temperature at which pyrolytic decomposition may occur, but is not heated to a temperature where it will actually begin to decompose in the fuel stream prior to entering the reactor.

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Once in the reactor, said fuel source is rapidly
heated to a temperature of between about 1000 and 2000°C
(when measured locally) for between 0.1 and 10 seconds by
interspersing said fuel source with air- or oxygen-
assisted hydrocarbon flames (produced by the hydrocarbons
of the flame fuel, which may be the same or different to
the fuel source) wherein the heat of the combustion
products causes localized heating resulting in pyrolytic
decomposition. Conveniently, the reactor is operated at
a pressure of between 0.1 and 150 bars.

The atmosphere in the reactor is composed solely of
the hydrocarbon fuel (including the flame fuel and fuel
source), the oxidizer (with any carrier gas) and the
combustion and/or decombustion and/or decomposition
products (e.g. solid carbon and hydrogen-rich gases). No
other compounds are included in the reactor.

In one of its simplest embodiments, the hydrocarbon
fuel is introduced into the reactor where it is
interspersed with oxygen and/or air in a controlled
manner and ignited thus causing local combustion limited
by the amount of oxygen which is introduced. The
periphery of the flames which intersperse with the
remainder of the hydrocarbon fuel contains the hot
combustion products which achieve temperatures in excess
of 1000°C thereby causing pyrolytic decomposition of
adjacent molecules of the hydrocarbon fuel. In this case
the flame fuel and feedstock fuel, which may be the same
or different, are portions of the same fuel (the
hydrocarbon fuel) which are mixed together.
Alternatively, the flame fuel and feedstock fuel, which
may be the same or different, may be separate from one
another and after ignition and combustion of the flame
fuel the combustion products are mixed with the feedstock
fuel to achieve temperatures in excess of 1000°C.

Due to the limited amount of oxygen which is
employed as oxidizer, the oxidizer is used up during the
combustion reaction. Thus, soot formed in the reaction
does not burn due to the absence of an oxidizer.

However, when using the combustion products to heat the
fuel source, oxygen may be produced by decomposition of
fractions of the combustion products H_2O and CO_2. To
avoid this, heating should be performed as rapidly as
possible as mentioned above to prevent burning of the
soot. This stimulates nucleus formation resulting in
soot formation.

As mentioned above, said hydrocarbon flame may be
produced by a hydrocarbon which is the same or different
to the hydrocarbon-containing feedstock, for example the
hydrocarbon of the flame may be acetylene. The
hydrocarbon/s of the hydrocarbon fuel may be selected to
alter the yield of solid carbon. For example, an
acetylene or hydrogen flame may be used in preference to
a methane flame due to the higher temperatures which the
former achieves which results in higher solid carbon
production.

Combustion products which are of a sufficient
temperature to heat the feedstock in the hydrocarbon fuel
may be produced in different ways. For example, a burner
system may be used which is associated with, optionally
external to, the reactor. Combustion in the burner
system may be achieved by reaction between a fuel (flame
fuel) specially introduced into the burner and an
oxidizer, for example oxygen from air to produce a flame.
Oxygen may be separated from the nitrogen in air and the
nitrogen itself may be used in conjunction with other
products of the methods described herein in chemical
processes.

Preferably, the burner or burner-like system
utilizes pure or nearly pure oxygen (with a moderate
admixture of nitrogen, preferably with a O:N
stoichiometric ratio greater than 1:1). The burner fuel
(equivalent to the flame fuel) which is used for
combustion may be the fuel corresponding to the feedstock
fuel, or a different fuel, or more preferably may be the

hydrogen-rich gas produced from the reaction of the
invention which is provided to the burner system via an
outlet of the reactor. The burner fuel which is used
must be able to achieve temperatures of 2000°C or more.
The use of the hydrogen-gas is preferred to avoid
excessive production of CO_2. Furthermore, reaction of
hydrogen with oxygen leads to very high temperatures.

Localized heating of the hydrocarbon-containing
feedstock fuel using the hot combustion products of the
burner may be achieved by mixing the combustion products
with the feedstock fuel such that rapid and temporary
heating (ie. 0.1 to 10 seconds) of the fuel occurs. The
C:O ratio referred to herein refers to the stoichiometric
ratio of all carbon and oxygen atoms involved in the
reaction (ie. from the oxidizer, hydrocarbon fuel and
carrier gases) and thus includes those involved in the
reaction in the flame. In the case in which a burner
system is used, the C:O ratio would hence also include
all carbon and oxygen atoms involved in the reactions in
the burner.

Flames which intersperse said hydrocarbon fuel or
feedstock fuel may be achieved by the use of a turbulent
flame. Such flames are effective due to their ability to
cause localized heating of the feedstock fuel, thus
generating the high temperatures at the flame/fuel
interface required for solid carbon formation. The
burner system is preferably designed in such a way that
the momentum of the combustion products (e.g. created by
using an oxygen-assisted flame) may be utilised to
perform a rapid mixing with the feedstock fuel (fuel
source). Similarly, the feedstock fuel may be brought
into contact with the combustion products by utilising
and enhancing the momentum of the feedstock fuel. To
achieve this aim, the interior of the reactor for
performance of the method of the invention may be
designed to allow counter flow, co-flow, cross flow (such
as in tangential reactors or axial reactors with radially

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introduced feedstock) or a combination thereof, of the
feedstock fuel and the hot combustion products of the
burner system. The properties of carbon black produced
may be varied by appropriate choice of the reactor and
the method of mixing of the combustion products and the
feedstock fuel. Appropriate reactors are described in
for example patent applications EP-A-360399 (Columbian
Chemicals), GB-1242391 (Phil Black Limited) and US-
3619140 (Morgan and Jordan).

In a counter-flow version of the invention, the
oxidizer, the flame fuel and/or the hot combustion
products from the burner system may be introduced through
nozzle-like inlet port at very high velocity (typically
20-200m/s), in an opposite direction to the other
components of the hydrocarbon fuel (ie. the feedstock
fuel), thus allowing the oxygen or hot combustion
products to penetrate into the feedstock fuel, creating
very hot fine structures, such that the formation of the
solid carbonaceous material takes place in a large part
of the feedstock fuel volume. The outlet of the reactor
should be arranged to optimize the interaction of the
feedstock fuel with the hot structures.

Alternatively, the oxidizer, the flame fuel and/or
the hot gases are introduced through nozzles which
surround the outlet, thus the feedstock fuel necessarily
encounters the hot flames.

In the above described cases, the feedstock fuel may
itself be introduced into the reactor in such a way to
form a vortex flow structure.

Both the co-flow and cross-flow versions of the
invention utilize the same mechanisms for their
production, namely creation of shear forces by the
interactions between the high velocity of one or more of
the oxidizer, combustion products, flame gas or other
components of the hydrocarbon fuel, ie. the feedstock
fuel to create inter-mixing of the heated structures in
the feedstock fuel. To achieve this effect, the inlet

ports may be designed in such a way to create cyclone-
like flow in the reactor.

In all cases, the mixing is designed such that rapid
mixing may be achieved. After mixing, the peak of high
local temperature disappears such that the temperature of
the product which leaves the reactor is homogenous and
preferably has a temperature in the order of 1000°C.

Alternatively, the interior of the reactor may be
designed in such a way that no burner is present, but the
hydrocarbon fuel and oxidizer are mixed and ignited, such
that part of the hydrocarbon fuel acts as the flame fuel
such that thermal reactions between the hydrocarbon fuel
and the oxygen takes place in a localized fashion leading
to temperature peaks exceeding 2000°C. One way to
achieve this is to create turbulent structures that will
contain the thermal reactions. The oxygen thereby
completely or nearly completely reacts to form CO_2 and
H_2O. The amount of carbon in the fuel converted to CO_2 is
small since the average heating of the hydrocarbon fuel
is moderate and of the magnitude of 1000°C, while the
reacting structures locally achieve a much higher
temperature and the amount of oxidizer which is available
is limited.

The above is preferably achieved by supplying the
oxidizer to the reactor in the form of pure oxygen or
nearly pure oxygen, but may also be achieved if the
oxygen includes a moderate admixture of nitrogen.

Thus, viewed from a preferred aspect, the present
invention provides a method of producing a solid
carbonaceous product wherein a hydrocarbon fuel
(comprising a bulk feedstock fuel and flame fuel) is
heated with oxygen at a C:O ratio greater than 1:3, e.g.
equal or greater than 1:2, said method comprising at
least the steps of pre-heating the bulk feedstock fuel
and passing said bulk feedstock fuel into a reactor in
which said bulk feedstock fuel is rapidly heated to a
temperature of between about 1000 and 2000°C for between

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0.1 and 10 seconds by interspersing said bulk feedstock fuel with air- or oxygen-assisted hydrocarbon flames utilizing said flame fuel, to obtain localized heating whereby to effect partial pyrolytic decomposition.

Preferably the bulk feedstock fuel and flame fuel are the same and are pre-heated together and this fuel is ignited to create the hydrocarbon flames.

The C:O ratio corresponds to the stoichiometric ratio of the total carbon and total oxygen admitted to the reactor (including those admitted to the burner which may be external to the reactor), wherein the latter may be in the form of O_2, H_2O, CO_2 etc. As mentioned above, the C:O ratio is preferably significantly greater than 1:2 and thereby the oxygen which leaves the reactor by the outlet is in the form of H_2O and most of the hydrogen is in the form of H_2.

The solid carbonaceous product produced by this method may be collected continually or intermittently during its production or at the end of the reaction. In cases where combustion of the hydrogen-rich gases which have been produced is to be performed, if this combustion is performed in a separate stage, the carbon may be collected prior to its initiation. Separation of the solid carbon may be performed by a cyclone system or filter-like or scrubber system, after which the solid carbon is cooled. However, if the production of solid carbon, hydrogen-rich gases and the latter's combustion is all performed sequentially or simultaneously in the same reactor, then the carbon black may be collected during or after the reaction.

The solid carbon thus formed has particular industrial applications and forms a further aspect of the invention. For example, the carbon may be used as an admixture in solid materials to modify the physical properties of the materials, such as their strength, thermal response, flexibility or wear.

The physical properties of the carbonaceous material

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may be modified by altering the operational conditions of
the reactor. For example, this may be achieved by
adjusting the inlet feeds to the reactor and/or reactor
pressure, or by modifying the interior structure of the
reactor such that the time scales of the heated
structures are changed. This may be achieved for example
by increasing the momentum of the oxidizer, flame gas,
fuel source and/or combustion products introduced into
the reactor. This may be achieved by increasing the flow
rate of one or more of the above components or by
reducing the input port or nozzle area and maintaining
the same flow rate.

The soot-like carbon may be formed into materials
such as carbon fibres, tubes, whiskers, spheres, shells
or to fill particles or used in the production of
materials such as printer's ink, technical rubber,
carbon-containing solid materials such as pipe-materials
and electrodes or as an additive such as to increase the
strength of other materials such as metal, concrete etc.

The above described method also produces hydrogen-
rich gas which is a combustible fuel which may be used as
an energy source.

The present invention additionally thus provides
a method of producing a hydrogen-rich gas with a H:C
ratio on atomic basis equal to or exceeding 20:1, (ie.
equal or less than 20% of the carbon in the feedstock
fuel is converted to CO_2, e.g. about 50:1) and a H:O ratio
greater or equal to 5:1 (e.g. about 10:1), wherein a
hydrocarbon fuel is heated with oxygen at a C:O ratio
greater than 1:3, preferably 1:2, especially preferably
greater than or equal to 1:0.2, to a temperature of at
least about 1000°C whereby to effect partial pyrolytic
decomposition of said fuel.

As mentioned above, the H:C, H:O and C:O ratios
referred to herein denote stoichiometric or atomic
ratios.

In the above method, the H_2O which is produced is

gaseous at the temperatures involved and is thus
considered to form part of the hydrogen-rich gas.

As mentioned in connection with the solid carbon,
this may be harvested during and/or after the reaction
which produces it.

The energy produced by combustion of the hydrogen-
rich gases of the invention may be used for any
conventional processes requiring energy. Thus for
example, combustion of the hydrogen-rich gases may be
linked to a gas turbine combustor, boiler, or other
machinery by appropriate modification of the reactor
pressure for connection with associated machinery. The
reactor and the interior of the reactor are preferably
designed and operated at pressures such that utilization
of the hydrogen-rich gas in energy systems may be readily
facilitated and not require compression of the gas before
it is used.

Such systems may have as their main purpose the use
of solid carbon produced according to the methods of the
invention, for example systems in which the solid carbon
is formed into pellets, blocks or as bulk material.
Alternatively, the systems may be designed to form
fullerences (pellets), diamonds or monodisperse spheres
of uniform size. The gas may also be used as fuel in a
furnace such as in steam raising furnaces, glass
furnaces, metallurgical furnaces or similar.
Advantageously, the above methods produce only small
amounts of CO_2 and offer environmentally friendly
alternatives to conventional processes.

The hydrogen-rich gas may also be used to provide
the fuel for the flame used in the method of the
invention. Alternatively, the heat of the gas exiting
from the reactor may be employed, for example to pre-heat
the fuel gas, thus allowing conservation of energy.

The invention thus further provides a method of
combustion wherein the hydrogen-rich gas produced
according to the invention is combusted. This combustion

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may be performed with an appropriate oxidizer (e.g. air
or oxygen) on the hydrogen-rich gas to produce energy
which may then be utilized or may form an integral part
of a chemical, metallurgical or similar process. To
reduce the production of undesirable combustion products,
and hence to reduce harmful emissions to the atmosphere,
oxygen-rich oxidizer (preferably pure or essentially pure
oxygen) is preferred.

Optionally, the energy evolved in the first stage of
the reaction, ie. pyrolytic decomposition and combustion
in the flame, may be utilized for the energy-requiring
processes contemplated above. It will be appreciated
that under certain circumstances, all or a part of the
energy evolved during the first or second stage of the
reaction may be recycled for use in initiating or
sustaining these reactions.

The hydrogen-rich gas may also be used in chemical
processes other than combustion to produce hydrogen-
containing chemicals. When the oxidizer used in the
above reaction is in the form of oxygen which has been
separated from nitrogen in air (as described above), the
nitrogen thus produced may be used in processes with the
hydrogen-rich gas produced above to produce nitrogen-
hydrogen compounds such as NH_3 or other compounds
including nitrogen and hydrogen and other elements. In
this respect, it will be appreciated that in reactions
described above, sufficient amounts of hydrogen-rich gas
are produced to ensure that nitrogen which is separated
from air to yield the oxygen which is used in the
reaction will be fully utilized to produce NH_3 with some
hydrogen-rich gas remaining.

Reactors appropriate for performance of the
invention form a further aspect of the invention.
Reactors of the invention comprise at least: one or more
inlet ports to carry the hydrocarbon fuel (or components
thereof) into the reactor cavity, through which the
oxidizer may also be carried, and one or more outlet

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ports, optionally with a capture chamber, through which evolved gases of the reactions may leave the reactor. Reactors are required to sustain temperatures required for performance of the above methods, ie. greater than about 2000°C and pressures of 0.1 to 150 bars and thus should be manufactured from heat resistant steel or similar and equipped with heat resistant internal cladding and insulation.

To allow ignition of the hydrocarbon fuel to produce flames which intersperse with the remaining hydrocarbon fuel (ie. the feedstock fuel), a source of ignition is provided which may be external to or within the reactor (or external to or within a chamber within the reactor).

In instances in which the flame (or its combustion products), flame fuel and/or the oxidizer is provided separately to the feedstock fuel, one or more further inlet ports may be provided, for example a burner system may be provided which produces a flame and allows combustion products to enter the reactor. In this way, combustion is completed prior to contact with the remaining feedstock.

Thus, viewed from a further aspect the present invention provides a reactor for performance of the methods of invention comprising at least: one or more inlet ports allowing entry of the hydrocarbon fuel into the reactor cavity, one or more outlet ports through which evolved gas may leave the reactor and a source of ignition whereby to effect partial combustion of hydrocarbons introduced into said reactor or associated burner.

Said burner may be external or internal to the reactor cavity or chamber. In both cases, combustion products enter said reactor chamber through one or more reactor inlet ports. In the case of an externally arranged burner, said burner is connected to said one or more inlet ports via one or more inlet tubes. Where a distinct reactor chamber is present in the reactor

cavity, appropriate inlet/outlet tubes or ports are
provided for communication across the space between the
reactor walls and the reactor chamber.

As mentioned previously, appropriate mixing of the
combustion products and the feedstock fuel may be
achieved by introducing the flame or combustion products
(if a burner is used), the flame gas, the oxidizer and/or
the hydrocarbon fuel or feedstock fuel through nozzle-
like inputs or ports at a velocity of 20-200m/s into
(preferably in an opposite direction) one or more of the
afore-mentioned fuels or gases, for example in an
opposite direction to the feedstock fuel. These nozzles
represent inlet ports and may replace or supplement
existing inlet ports. They may be arranged in various
positions within the reactor, or arranged in appropriate
patterns, for example around the reactor outlet. Thus,
the use of multiple inlet ports for the entry of
feedstock fuel, flames, combustion products, flame gas,
and/or oxidizer, of a diameter suitable to allow
interspersion with other elements of the reaction when
operated at an appropriate flow rate, is preferred.

Preferred embodiments of the invention will now be
described with reference to the Figures in which:

Figure 1 shows a reactor for use according to the
invention in which the oxidizer is admitted to the
reactor through a separate inlet port;

Figure 2 shows a reactor for use according to the
invention in which internal heating is provided by an
integral burner system with separate flame fuel and
oxidizer supplies in which the fuel may be the same as
the feedstock fuel or provided by the hydrogen-rich gas
which is produced or may be a separate fuel source;

Figure 3 shows a reactor similar to the reactor
shown in Figure 2 but with an external burner system;

Figure 4 shows a reactor fitted with a simple
control system; and

Figure 5 shows how the reactors may be coupled to

energy or gas turbine systems.

Figure 1 shows a reactor with an internal reactor
chamber (1) within the reactor. The reactor additionally
comprises a fuel inlet port (2), an oxidizer inlet port
(3) and an outlet port (4). These inlet ports are
appropriately connected to the reactor chamber (1) to
allow entry of the media carried through the inlet/outlet
ports, e.g by reactor chamber inlet ports (not shown) or
by the use of a chamber which is not sealed, e.g. a
cylinder-shaped chamber with unsealed ends. The reactor
chamber may be associated with the reactor cavity inner
walls or may be separate therefrom allowing the passage
of gases/solid between the reactor cavity inner wall and
the reactor chamber outer wall, e.g via inlet/outlet
tubes or ports.

A source of ignition (5) is provided to ignite
hydrocarbons in the reactor at an ignition zone and may
be positioned exterior of or within the reactor chamber.
A heat source (6) is arranged in sufficient proximity to
the fuel inlet port (2) to allow heating of the
hydrocarbon fuel. The reactor is insulated (7) and heat
recovery may be achieved in an appropriate manner using
an energy capture means (8), such as by heating H_2O
containing structures or other heat absorbing medium such
as oil, liquid metal, gas, air or other appropriate
substance. A further outlet port (9) may be provided, in
this case exterior to the reactor chamber, for
collection of the solid carbon material by use of a
carbon removing device (10) such as a cyclone, filter,
scrubber, scraper or electrical/mechanical precipitator.

In this embodiment, hydrocarbon fuel (containing the
feedstock and flame fuel) is passed into the reactor
through inlet port (2) after it has been pre-heated by
the heating means (6). The pre-heating means may be
provided by the outlet gas from the reactor. Oxidizer
then enters the reactor and chamber via inlet port (3)
and mixes with the hydrocarbon fuel to produce a

combustible mixture. This is ignited by the ignition
means (5) resulting in combustion to the extent that the
presence of the oxidizer allows.

The hot combustion products are then interspersed as
described previously by the action of shear generated
turbulence at the interface between the hydrocarbon fuel
and the high velocity oxidizer (e.g. oxygen). This
allows hot combustion products or flames into the areas
of the fuel gas which have not been combusted to produce
thermal reactions at the interface of the combustion
product and the unspent fuel leading to the production of
solid carbon.

This carbon may be collected by the carbon removing
device (10) and collected through the outlet port (9).
Hydrogen-rich gas leaves the reactor through outlet port
(4) and may be utilized for heating the hydrocarbon fuel
or in a different energy requiring system. Energy
produced (in the form of heat) in the reactor during the
combustion and decomposition processes may be collected
through appropriate energy capture means (8).

In Figure 2, the oxidizer inlet port (3) and source
of ignition (5) of Figure 1 is replaced with a burner
(11) which is fed via an inlet port (12) which carries
the oxidizer and flame gas to the burner for ignition.
The burner includes an ignition means. The flame gas may
be the same as the feedstock fuel, or may be the
hydrogen-rich gas produced in the reactor or a separate
fuel source. Similarly, in Figure 3, the burner (11) may
be placed external to the reactor, and be connected
thereto by an inlet port (3) and the flame fuel and
oxidizer provided into the burner via inlet port (12).

Figure 4 shows a schematic representation of the
reactors shown in Figures 1 to 3 in which various control
valves are included. Thus, entry of the fuel may be
controlled by flow control valve (1) into the reactor and
by flow control valve (2) into a burner or point of
ignition. Flow control valve (3) may be provided to

control the flow of oxidizer into the reactor. Thus the
reactor as described above may be made controllable in
such a way that the composition of the products may be
altered continuously and controlled according to
operating procedures and product specifications. This
advantageously allows variable emission control and the
ability to achieve optimum power at such emission levels.

As will be appreciated from the discussion herein,
the amount of oxidizer which is introduced into the
reactor is crucial and thus this valve is. of particularly
importance for controlling the reaction. Pressure
control valve (4) may be used to control the release of
the gas which is produced in the reactor. Filter (5)
onto which the solid carbon is collected may be linked to
the outlet port carrying the hydrogen-rich gas and the
flow of carrier gas from the filter may then be
controlled by a control valve (6).

Figure 5 shows how the energy produced in the 2-
stage process of the invention may be used to power a gas
turbine. In this case feedstock fuel is passed via port
(1) into the reactor (2) and heated by the hot combustion
products which enter the reactor at inlet port (3)
produced by the combustion of a flame fuel and an
oxidizer. The hydrogen-rich gas which exits at outlet
port (4) is passed to a burner (5) where in the presence
of air or oxygen, which is compressed and provided via
burner inlet port (5a), it is combusted. The combustion
products exit the burner via outlet port (6) and are used
to power a turbine (7) which in turn is used to generate
electricity.

To optimise the efficiency of this process, and thus
to reduce environmental impact, side-products of the
reaction may be utilized. Appropriate heat-exchange
mechanisms may be employed, for example at (8) as shown
in the previous Figures, at (9) to warm air to be passed
into the burner and at (10) to capture the heat of
exhaust gases. As indicated on Figure 5, the heat of the

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exhaust gases may be used to heat the oxidizer to be used
in the burner (5). Figure 5 also illustrates how energy
produced in the 2-stage process may be used for boiler
driven power plants (alternative 2). In that case, the
combustion products which exit the burner via outlet port
(6) are used to drive a boiler (11). Again appropriate
heat exchange mechanisms may be used to improve
efficiency.

Use of the above reactors for the production of the
solid carbon or hydrogen-rich gas or for the production
of energy form further aspects of the invention.